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(54) Title: UV VISIBLE/DAYLIGHT INVISIBLE FLUORESCENT PIGMENT**(57) Abstract**

Compositions evidencing ultraviolet radiation stimulated blue fluorescence when combined with compositions evidencing ultraviolet radiation stimulated yellow fluorescence when viewed in daylight are colorless but manifest an intense white fluorescence under ultraviolet radiation. The resultant product is a heavy metal free fluorescent product of interest for use in printing inks, functional displays and for decorative purposes.

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UV VISIBLE/DAYLIGHT INVISIBLE FLUORESCENT PIGMENT

Field of the Invention

This invention relates to a fluorescent pigment or colorant formulation. More particularly,
5 the present invention relates to a fluorescent colorant formulation comprising two or more fluorescent compositions wherein the sum of the fluorescent emissions produces novel visual/optical effects when stimulated by ultraviolet radiation.

10

Background of the Invention

Fluorescent or phosphorescent colorant formulations and pigments have been commonly used,
15 imprinted, coated upon or otherwise incorporated into objects, packaging materials or documents for the purpose of producing markings such as codings, patterns or designs and images which although invisible in daylight can be perceived or registered under
20 ultraviolet radiation.

Fluorescent inks such as solvent based screen printing inks typically comprise a three component system wherein a fluorescent dye is dissolved in a matrix which is dispersed in a vehicle. The resulting
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compositions exhibit characteristics formulated for specific applications such as lithographic or letter-press printing.

5 Water base paints have also been commonly prepared by combining pigments and dyes which are phosphorescent and appear transparent or colorless under visible light. These compositions typically include heavy metal additives which produce white light
10 when stimulated by ultraviolet radiation.

15 In recent years, workers in the art have focused their interest upon this technology with a view toward lessening the environmental impact of such compositions which tend to be more toxic when employed
in high concentrations.

Brief Description of the Invention

20 In accordance with the present invention, the prior art limitations have been overcome by means of a novel fluorescent pigment or color formulation comprising two or more fluorescent components, such as pigments or dyes, in which the sum of the fluorescent
25 emissions produces novel visual and optical effects when stimulated by ultraviolet radiation. More specifically, it has been found that a composition which produces ultraviolet radiation stimulated blue fluorescence when combined with a composition which
30 produces ultraviolet stimulated yellow fluorescence,

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each of which is normally transparent or colorless when viewed in daylight, produces an intense white fluorescence under ultraviolet radiation. The result is a heavy metal-free fluorescent product which is of
5 use in printing inks for security applications, tracking or sorting, perception enhancement, tagging and identification, sensor readouts, visual enhancement of indicators, functional displays and for decoration/entertainment purposes or when prepared in a
10 suitable vehicle may serve in other coating or coloring applications such as in paints or plastic colorants.

Ultraviolet fluorescent or phosphorescent substances incorporated in colorant formulations to produce the desired optical effects described herein
15 may be selected from among coumarins, benzotriazoles, oxazinones, quinazolinones, benzopyranones, unsaturated biphenyl derivatives, organic phosphors and the like. It has been found that these compositions may be
20 incorporated directly into a color formulation in proportions designed to yield the desired optical effect, or alternatively, colorants having comparable optical effects may be produced by first preparing pigments containing individual ultraviolet fluorescent
25 or phosphorescent components and then combining such pigments in suitable proportions.

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DETAILED DESCRIPTION OF THE INVENTION

The first step in the practice of the present invention involves selection of a resin suitable for use in the practice of the present invention. Resins suitable for this purpose include the well known formaldehyde-aminotriazine-sulfonamide (FTS) matrix, polyurethane, polyester or polyester-polyamide resins.

10 The colorants or pigments selected for use in the practice of the present invention are employed in proportions designed to produce the desired optical effects. Studies have revealed that it is advantageous to employ concentrations of blue fluorescent pigment
15 and yellow fluorescent pigment wherein the sum of the blue and yellow components ranges from 10% to 95% based upon the weight of composition of interest.

20 The novel optical effects achieved herein are attained by combining in the correct proportions two or more components which under ultraviolet radiation fluoresce or phosphoresce blue and yellow, respectively but when combined yield an emission which appears white to the observer but registers as two distinct visible
25 emissions by an appropriate sensing device. These colorant formulations appear transparent or colorless under visible light (4,000 to 7,000 nanometers) but yield the desired visible optical effects when stimulated by ultraviolet light having a wavelength of
30 less than 4,000 nanometers.

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Conventional additives such as dispersing agents, antioxidants, ultraviolet inhibitors, driers and the like may also be present. Specific visual effects may be created by the addition of other
5 pigments.

The resultant compositions may be used as prepared or may be modified with additional solvents or additives to achieve specific properties.

10

The following examples of the practice of the present invention have been set forth to more fully demonstrate the invention. It will be appreciated by those skilled in the art that these examples are set forth solely for purposes of exposition and are not to be construed as limiting.
15

Example 1

20

This example describes the preparation of an ultraviolet-white/daylight invisible water base paint.

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Part 1A. UV-Blue Water Base Paint Concentrate

	<u>Composition</u>	<u>Weight%</u>
	Water	40.0
5	Hydroxyethyl cellulose	0.8
	anionic dispersing agent	2.0
	non-ionic dispersing agent	0.2
	UV-fluorescent blue pigment	54.0
	defoamer	0.2
10	propylene glycol	<u>2.8</u>
		100.0

The foregoing compositions were added to a reaction vessel in the order listed and mixing initiated at a mixing speed of 1,500 rpm for a period of 5 minutes in a conventional prior art mixer. The resultant product was a UV-blue concentrate dispersion.

Part 1B UV-Blue Water Base Paint

	<u>Composition</u>	<u>Weight%</u>
20	2% Hydroxyethyl cellulose in aqueous solution	21.0
	propylene glycol	2.1
	Part 1A UV-blue paint concentrate	43.0
25	latex	<u>33.9</u>
		100.0

The foregoing materials were added to a reaction vessel in the order listed and mixing initiated for a time period of 5 minutes at 1,500 rpm in a conventional

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prior art mixer. The resultant product was a UV-blue paint.

Part 1C UV-Yellow Water Base Paint

5

	<u>Composition</u>	<u>Weight%</u>
	2% Hydroxyethyl cellulose in aqueous solution	40.0
	propylene glycol	2.0
10	UV-fluorescent yellow pigment dispersion	20.7
	latex	32.4
	water	<u>4.9</u>
		100.00

15 The foregoing materials were added to a reaction vessel in the order listed and mixing initiated for a time period of 5 minutes at 1,500 rpm in a conventional prior art mixer. The resultant product was a UV-yellow paint.

20

Part 1D UV-White Daylight Invisible Water Base Paint

25

	<u>Composition</u>	<u>Weight% Range</u>
	Part 1B UV-blue paint	72.0 to 88.0
	Part 1C UV-yellow paint	<u>28.0 to 12.0</u>
		100.00

30 The foregoing materials were added to a reaction vessel in the order listed and mixing initiated for a time

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period of 5 minutes at 1,500 rpm in a conventional prior art mixer. The resultant product was a UV-white paint.

5 **Example 2**

This example describes the preparation of an ultraviolet-white/daylight white solvent white screen printing ink.

10

Part 2A. Screen Printing Ink Base

	<u>Composition</u>	<u>Weight%</u>
15	9/6% solution of ethylhydroxyethyl cellulose	44.0
	<u>Material</u>	<u>Weight%</u>
	modified polysaccharide	9.6
	Stoddard solvent	86.8
20	primary amyl alcohol	<u>3.6</u>
		100.0
	vinyl toluene copolymer	19.4
	70% resin in petroleum hydrocarbon solvent (Nevpene 9500 in Stoddard solvent)	24.6
25	solvent borne organic ink	7.6
	antioxidant	0.6
	solvent	3.1
	2-ethylhexanol	<u>0.7</u>
30		100.0

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The foregoing materials were added to a reaction vessel in the order listed and mixed for a time period of 5 minutes at 1,500 rpm.

5 **Part 2B UV-Blue Screen Printing Ink**

	<u>Composition</u>	<u>Weight%</u>
	Part 2A Screen Printing ink base	55.0
10	UV-fluorescent blue pigment	<u>45.0</u>
		100.0

The foregoing materials were added to a reaction vessel in the order listed and mixed for a time period of 5 minutes at 1,500 rpm to produce a UV-blue solvent-based screen.

Part 2C UV-Yellow Screen Printing Ink

20	<u>Composition</u>	<u>Weight%</u>
	Part 2A Screen printing ink base	95.0
	UV-fluorescent yellow pigment	<u>5.0</u>
		100.0

25 The foregoing materials were added to a reaction vessel in the order listed and mixed for a time period of 5 minutes at 1,500 rpm to produce a UV-yellow solvent-based screen printing ink.

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Part 2D Daylight White Screen Printing Ink

	<u>Composition</u>	<u>Weight%</u>
5	Part 2A screen printing ink base	95.0
	Rutile titanium dioxide	5.0
		100.0

10 The foregoing materials were added to a reaction vessel
in the order listed and mixed for a time period of 5
minutes at 1,500 rpm to produce a white solvent-based
screen printing ink.

15 **Part 2E UV White/Daylight White Solvent Base Screen
Printing Ink**

	<u>Composition</u>	<u>Weight% *</u>
20	Part 2B UV-Blue Screen Printing Ink	28.9 to 35.2
	Part 2C UV-Yellow Screen printing ink	62.1 to 55.8
25	Part 2D White Screen Printing Ink	<u>9.0</u> 100.00

* the sum of the blue and yellow component in weight
per cent was 91.00%.

30 The foregoing materials were added to a reaction vessel
in the order listed and mixed for a time period of 5

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minutes at 1,500 rpm to produce a UV fluorescent white, solvent-based screen white printing ink.

Example 3

5

This example describes the preparation of a UV-white/daylight invisible plastic colorant.

Part 3A UV-White Plastic Colorant Concentrate

10

<u>Composition</u>	<u>Weight %*</u>
UV fluorescent blue pigment	7.47 to 9.13
UV fluorescent yellow pigment	2.53 to 0.87
low density polyethylene powder	40.0
low density polyethylene pellets	<u>50.0</u>
	100.0

* the sum of the blue and yellow component in weight per cent was 10.00%.

The foregoing materials were combined and thoroughly mixed for 5 minutes by means of a paint shaker. The resulting mixture was compounded into pellets using a single screw extruder set at 400°F.

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Part 3B Production of UV White/Daylight Invisible Colored Plastic

<u>Composition</u>	<u>Weight%</u>
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5	High density polyethylene pellets	80.00
	Part 3A pelletized concentrate	<u>20.00</u>
		100.00

10 The foregoing materials were combined and thoroughly mixed and then fed into an injection molder and molded into chips at 400 per °F. The resulting chips appeared uncolored under visible light and fluoresced white under ultraviolet light.

15

Example 4

This example describes the preparation of a UV-white/daylight invisible lithographic ink.

20

<u>Composition</u>	<u>Weight% Range*</u>
--------------------	-----------------------

UV-fluorescent blue lithographic vehicle	38.25 to 46.75
pigment dispersion	
lithographic vehicle	28.5
quaternary amine derivative of fatty acid	5.0
condensate	
UV-fluorescent yellow pigment	24.25 to 15.75
Ink oil	<u>4.0</u>
	100.00

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* the sum of the blue and yellow component in weight per cent is 62.50%.

The UV-fluorescent blue lithographic vehicle
5 pigment dispersion, the lithographic vehicle, the
quaternary amine derivative and the ink oil were loaded
into a suitable mixing vessel. Mixing was then
effected at low speed (500-1,500 rpm) until homogeneity
was attained. Then, the UV fluorescent yellow pigment
10 was added and mixing continued until it was well
dispersed. It should be noted that the amount and
types of lithographic vehicle and ink oils can be
altered to achieve specific levels of tack and
viscosity in the ink.

15

Example 5

This example described the preparation of a
UV-White Daylight Invisible Flexographic/Gravure Ink.

20

<u>Composition</u>	<u>Weight % Range*</u>
Acrylic resin solution	33.7
Acrylic polymer emulsion	33.7
Associative thickener	2.7
Surfactant	1.4
Optical brightener	0.63 to 0.77
UV-fluorescent yellow pigment	4.57 to 4.43
n-propanol	8.0

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Water	15.0
Defoamer	<u>0.3</u>
	100.0

5 * The sum of the blue and yellow components is 5.20%

The acrylic resin solution and the acrylic polymer emulsion were loaded into a suitable mixing vessel and mixing effected at a low speed (500-800 rpm) 10 until homogeneity was attained. Then, the associative thickener, surfactant, one-half of the water and the defoamer were added and mixing continued until homogeneity was attained. Then, the optical brightener and the UV-fluorescent yellow pigment were added and 15 mixing continued for a time period ranging from 15-20 minutes. Following, the remaining water was pre-mixed with the propanol and the mixture added to the batch and stirred until homogeneous.

20 **Example 6**

This example describes the preparation of a UV-White Daylight Invisible Solvent Base Bulletin Paint

25 **Part 6A UV-Blue Solvent Base Bulletin Paint**

<u>Composition</u>	<u>Weight%</u>
Acrylic resin solution	47.0
UV -fluorescent blue pigment	35.0

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Solvent	<u>18.0</u>
	100.0

5 The acrylic resin and UV-fluorescent blue pigment were loaded into a suitable mixing vessel and mixed at low speed (1500rpm) for 5 minutes to produce a uniform dispersion. The solvent was then added and stirring continued an additional 5 minutes to yield a UV-fluorescent blue solvent base bulletin paint.

10

Part 6B UV-Yellow Solvent Base Bulletin Paint

<u>COMPOSITION</u>	<u>Weight%</u>
Acrylic resin solution	47.0
UV-fluorescent yellow pigment	35.0
Solvent	<u>18.0</u>
	100.0

20 The acrylic resin and UV-fluorescent yellow pigment were loaded into a suitable mixing vessel and mixed at low speed (1500rpm) for 5 minutes to produce a uniform dispersion. The solvent was then added and stirring continued an additional 5 minutes to yield a UV-fluorescent yellow solvent base bulletin paint.

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Part 6C UV-White/Daylight Invisible Solvent Base

Bulletin Paint

	<u>Composition</u>	<u>Weight% Range</u>
5	Part 6A UV-Blue Solvent Base Bulletin Paint	9.0 to 11.0
	Part 6B UV-Yellow Solvent Base Bulletin Paint	<u>91.0 to 89.0</u>
		100.0

10

The materials were loaded into a suitable mixing vessel in the order noted and mixed at low speed (1500rpm) for 5 minutes to produce a UV-white uniform dispersion. The solvent was then added and stirring continued an additional 5 minutes to yield a UV-white daylight invisible solvent base bulletin paint.

Example 7

20

This example describes the preparation of a UV-White Daylight Invisible Solvent Base Alkyd Paint

Part 7A UV-Blue Solvent Base Alkyd Paint

25	<u>Composition</u>	<u>Weight%</u>
	Modified alkyd resin solution	54.6
	UV-blue luminescent pigment	9.1
	Xylene	<u>36.3</u>
		100.0

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The modified alkyd resin solution and UV-fluorescent blue luminescent pigment were loaded into a suitable mixing vessel and mixed at low speed (1500rpm) for 5 minutes to produce a uniform
5 dispersion. The xylene was then added and stirring continued an additional 5 minutes to yield a UV-fluorescent blue solvent base alkyd paint.

Part 7B UV-Yellow Solvent Base Alkyd Paint

10

	<u>Composition</u>	<u>Weight%</u>
	Modified alkyd resin solution	54.6
	UV-blue luminescent pigment	9.1
15	Xylene	<u>36.3</u>
		100.0

The modified alkyd resin solution and UV-fluorescent yellow luminescent pigment were loaded
20 into a suitable mixing vessel and mixed at low speed (1500rpm) for 5 minutes to produce a uniform dispersion. The xylene was then added and stirring continued an additional 5 minutes to yield a UV-fluorescent yellow solvent based alkyd paint.

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Part 7C UV-White/Daylight Invisible Solvent Base Alkyd
Paint

	<u>Composition</u>	<u>Weight% Range</u>
5	Part 6A UV-Blue Solvent Base Alkyd Paint	94.5 to 95.5
	Part 6B UV-Yellow Solvent Base Alkyd Paint	<u>5.5 to 4.5</u>
10		100.0

The materials were loaded into a suitable mixing vessel in the order listed and mixed at low speed (1500rpm) for 5 minutes to produce a UV-white uniform dispersion. The solvent was then added and stirring continued an additional 5 minutes to yield a UV-white daylight invisible solvent base bulletin paint.

20 While the invention has been described in detail in the foregoing specification and the exemplary embodiments, it will be appreciated by those skilled in the art that variations may be made without departing from the spirit and scope of the invention.

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What is claimed is:

1 1. UV visible/daylight invisible colorant composition
2 comprising an ultraviolet radiation stimulated blue
3 fluorescent composition and an ultraviolet radiation
4 stimulated yellow fluorescent composition, the sum of
5 fluorescent emissions under ultraviolet radiation being
6 white fluorescence.

1 2. Water base paint composition in accordance with
2 claim 1.

1 3. Water base paint composition in accordance with
2 claim 2 wherein the colorant composition comprises from
3 72.0-88.0 weight per cent ultraviolet radiation
4 stimulated blue fluorescent paint and from 28.0 to 12.0
5 weight per cent ultraviolet radiation stimulated yellow
6 fluorescent paint.

1 4. Water base paint composition in accordance with
2 claim 2 wherein the colorants are selected from the
3 group consisting of coumarins, benzotriazoles,
4 oxazinones, quinazolinones, benzopyranones, unsaturated
5 biphenyls and organic phosphors.

1 5. Composition in accordance with claim 1 further
2 comprising a surfactant.

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1 6. UV visible/daylight invisible solvent base screen
2 printing ink including an ultraviolet radiation
3 stimulated blue screen printing ink, an ultraviolet
4 stimulated yellow screen printing ink and a white
5 screen printing ink.

1 7. Screen printing ink in accordance with claim 6
2 comprising from 28.1 to 35.2 weight per cent
3 ultraviolet radiation stimulated blue screen printing
4 ink, from 62.1 to 55.8 ultraviolet radiation stimulated
5 yellow screen printing ink and 9.0 weight per cent
6 white screen printing ink.

1 8. Ultraviolet radiation stimulated white plastic
2 colorant concentrate comprising an ultraviolet
3 radiation fluorescent blue pigment, an ultraviolet
4 radiation fluorescent yellow pigment, low density
5 polyethylene powder and low density polyethylene
6 pellets, the sum of the blue and yellow pigments being
7 10.0 weight per cent based upon the weight of the total
8 composition.

1 9. Ultraviolet radiation stimulated white
2 lithographic ink comprising an ultraviolet fluorescent
3 blue lithographic vehicle pigment dispersion, a
4 lithographic vehicle, a quaternary amine derivative of
5 a fatty acid condensate, an ultraviolet radiation
6 stimulated yellow pigment and an ink oil, the sum of
7 the blue and yellow components being 62.5 weight per
8 cent based upon the weight of the total composition.

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1 10. Ultraviolet radiation stimulated white
2 lithographic ink in accordance with claim 9 comprising
3 38.25 to 46.75 weight per cent ultraviolet fluorescent
4 blue lithographic vehicle pigment dispersion, 28.5
5 weight per cent lithographic vehicle, 5.0 weight per
6 cent quaternary amine derivative of a fatty acid
7 condensate, from 24.25 to 15.75 weight per cent
8 ultraviolet radiation stimulated fluorescent yellow
9 pigment, remainder ink oil.

1 11. Ultraviolet radiation stimulated white solvent
2 base bulletin paint comprising from 9.0 to 11.0 weight
3 per cent ultraviolet radiation stimulated blue solvent
4 base bulletin paint and from 91.0 to 89.0 weight per
5 cent ultraviolet radiation stimulated yellow solvent
6 base bulletin paint.

1 12. Ultraviolet radiation stimulated white solvent
2 base alkyd paint comprising an ultraviolet radiation
3 stimulated blue solvent base alkyd paint and an
4 ultraviolet radiation stimulated yellow solvent base
5 alkyd paint.

1 13. White solvent base alkyd paint in accordance with
2 claim 12 comprising from 94.5 to 95.5 weight per cent
3 blue solvent base alkyd paint, remainder yellow solvent
4 base alkyd paint.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/05046

A. CLASSIFICATION OF SUBJECT MATTER	IPC 6	C09B67/22	C09B67/20	C09D11/00	C09D5/02	C09K11/06
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According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C09B C09D C09K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 42 12 170 A (SAMSUNG ELECTRON DEVICES CO. LTD.) 25 February 1993 see column 1, line 1 - line 18 see column 1, line 55 - line 61 see examples ---	1
X	EP 0 137 201 A (KABUSHIKI KAISHA TOSHIBA) 17 April 1985 see page 3, line 23 - page 4, line 23 see examples ---	1
X	EP 0 091 184 A (KASEI OPTONIX LTD.) 12 October 1983 see examples 15-18 ---	1

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Int'l. Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Week 8211 Derwent Publications Ltd., London, GB; AN 82-20529E'11! XP002106050 "Mixed fluorescent composition" & JP 57 021483 A (MITSUBISHI ELECTRIC CORPORATION), 4 February 1982 see abstract ----	1
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Information on patent family members

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